16-μm infrared generation by difference-frequency mixing in diffusion-bonded-stacked GaAs

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Tunable 90-ps 15.6–17.6-μm coherent radiation was generated by means of difference-frequency mixing in diffusion-bonded-stacked GaAs. The sample consisted of 24 alternately rotated layers with a total length of 6 mm and with low optical loss to achieve third-order quasi-phase matching. The wavelength-tuning curve was close to the theoretical prediction, demonstrating that the bonding process maintained nonlinear optical phase matching over the entire interaction length. Maximum conversion efficiency of 0.7%, or 5% internal quantum efficiency, was measured at 16.6 μm, consistent with the theoretical predictions. © 1998 Optical Society of America

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The use of a stack of alternately rotated diffusion-bonded GaAs plates as a quasi-phase-matching structure was suggested by Gordon *et al.* in 1993.[1] Wafer fusion creates a monolithic body with periodic change in the nonlinear coefficient, eliminating the Fresnel reflections and scattering at the interfaces that limit air-spaced stacks of rotated plates. This technique transforms single-crystal GaAs into a phase-matchable synthetic nonlinear optical crystal. The resulting diffusion-bonded-stacked (DBS) GaAs maintains the advantages of single-crystal GaAs: a large nonlinear coefficient, good optical transmission from 2 to 16 μm, high thermal conductivity and optical damage threshold, good chemical stability and mechanical properties, and a well-developed growth technology at low cost. Noncritical phase matching with DBS GaAs has a wider angular acceptance and is easier to align than the earlier approach using a stack of GaAs plates at Brewster angle incidence.[2] DBS GaAs does not experience Poynting vector walk-off, permitting tighter focusing over a longer interaction length. DBS GaAs also has a large theoretical wavelength, 0.5 μm, and temperature acceptance, 270 °C (FWHM), for frequency doubling of 10.6-μm CO_2 laser radiation with a 1-cm-long crystal.

However, reducing the optical loss of DBS GaAs to that of single-crystal GaAs is a major challenge. Low optical loss not only increases energy-conversion efficiency in nonlinear interactions but also is essential for scaling to high average powers and for resonant devices. In this Letter we report optical device results with recently fabricated multilayer (>20-layer) stacks with low optical losses.

GaAs wafer bonding for nonlinear optics applications is discussed in Ref. 3. Undoped semi-insulating {100} double-side polished wafers (American X-tal Technology) were diced into 1-cm squares, stacked together, and put into a bonding furnace. The furnace was pumped to 30 mTorr and refilled with H_2 at temperatures below 600 °C. A pressure of 40 kg/cm^2 was applied at 600 °C, and the pressure in the furnace was maintained at 30 mTorr of H_2. The furnace was then refilled with H_2 to 10 psi (gauge) and kept at 676 °C for 4 h.

In this Letter we report a nonlinear interaction in a 24-layer DBS GaAs device with an average layer thickness of 252 μm. We fabricated the GaAs stack for third-order quasi-phase-matched difference-frequency generation (DFG) of 16.6-μm radiation by mixing 4.79- and 6.74-μm inputs from a $ZnGeP_2$ (ZGP) optical parametric generator (OPG) pumped by a 2.8-μm Er:Cr:YSGG laser.

Figure 1 shows the experimental setup for DFG. A double-pass ZGP OPG pumped by single pulses of a 2.8-μm actively mode-locked, *Q*-switched, cavity-dumped Er:Cr:YSGG laser generated 90-ps 4.79- and 6.74-μm input radiation with orthogonal polarization for the DFG process at a 3-Hz repetition rate, with a linewidth of 10–15 cm^{-1}.[4] We used type II phase matching to obtain narrow linewidth. An InSb wafer at normal incidence after the DBS GaAs crystal filtered out input radiation at wavelengths shorter than 7 μm to allow detection of the 16.6-μm output. A mercury cadmium telluride photoconductive detector with 25-μm long-wave cutoff at 77 K was used for detection.

The energy-conversion efficiency and the wavelength-tuning curve are both affected by the optical loss of the bonded stack. Figure 2 shows the optical absorption of the 6-mm bonded stack, measured with a Bio-Rad Model FTS-40 Fourier transform infrared spectrometer with a 1-cm-long GaAs single crystal as reference. Single-crystal GaAs has a measured optical loss of less than 0.1 cm^{-1} from 2 to 12 μm. The optical loss of the bonded stack is almost the same as that of single-crystal GaAs at long wavelengths and

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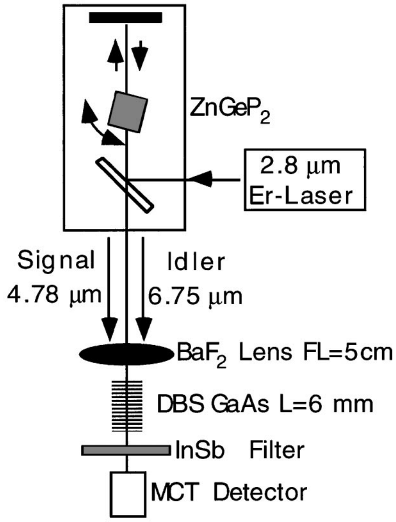

Fig. 1. Experimental apparatus for DFG with DBS GaAs. A double-pass type II phase-matched ZGP OPG pumped by single pulses of a 2.8-μm mode-locked, Q-switched Er:Cr:YSGG laser generated input radiation for the DFG process. The InSb wafer filters out input radiation. A liquid-nitrogen-cooled mercury cadmium telluride (MCT) photoconductive detector detects the DFG output.

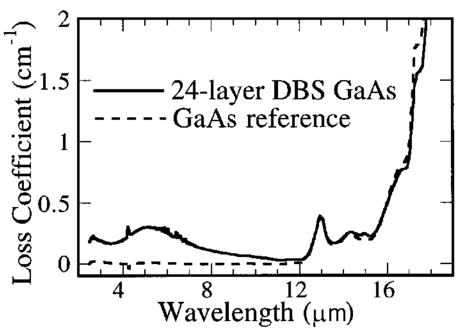

Fig. 2. Optical loss of the 24-layer DBS GaAs device. It has an average layer thickness of 252 μm for third-order quasi-phase-matched DFG of 16.6-μm radiation by means of mixing 4.79- and 6.74-μm inputs. The optical absorption of a 1-cm-long GaAs single crystal is used as a reference.

less than 0.3 cm^{-1} at short wavelengths. This optical loss is more than five times less than that of our first multiple-layer stack bonded 4 years ago. Since the generated 16.6-μm radiation is at the long-wavelength absorption edge of GaAs, the optical absorption is not negligible, even in the single-crystal sample.

The conversion efficiency and the phase-matching condition in the presence of loss can be calculated in the small-signal approximation, where the pump depletion that is due to DFG and the signal gain are neglected.

Let $E_p(z) = E_{p0}\exp(-\alpha_p z/2)$ and $E_s(z) = E_{s0}\exp(-\alpha_s z/2)$, where subscripts p, s, and i denote DFG pump, signal, and idler (DFG output), such that $\omega_p > \omega_s > \omega_i$, and α is the optical power loss. Under these assumptions, the idler field is given by[5] $\mathrm{d}E_i/\mathrm{d}z + \alpha_i E_i/2 = ik_i E_p E_s^{*}\exp(i\Delta kz)$, which gives

$$\frac{I_i}{I_s(0)} = \frac{\omega_i}{\omega_s}\Gamma^2 L^2 \frac{\exp(-\alpha_i L)}{(\Delta\alpha L/2)^2 + (\Delta kL)^2} \times \left\{\left[1 - \exp\left(-\frac{\Delta\alpha L}{2}\right)\right]^2 + 4\exp\left(-\frac{\Delta\alpha L}{2}\right)\sin^2\left(\frac{\Delta kL}{2}\right)\right\},$$

where

$$\Gamma^2 = \frac{2\omega_i\omega_s}{n^3c^3\epsilon_0}{d_{\mathrm{eff}}}^2 I_p\,,$$

where I is intensity, L is crystal length, $\Delta\alpha = \alpha_s + \alpha_p - \alpha_i$, and $\Delta k = k_p - k_s - k_i$; d_{eff} is the effective nonlinear coefficient.

For a lossless medium, $\alpha_i = \Delta\alpha = 0$, the nonlinear conversion reduces to the expected value of $I_i/[I_s(0)] = \omega_i/\omega_s\Gamma^2L^2\,\mathrm{sinc}^2(\Delta kL/2)$, which is the ideal conversion efficiency.

Figure 3 shows the effective nonlinear coefficient d_{eff} versus the azimuth angle for two input polarization states. In one case, as shown in Fig. 1, the polarizations of the two input beams were orthogonal, and one polarization was along the [110] axis of the GaAs. This configuration results in a maximum $d_{\mathrm{eff}}(\max) = (1/3)(2/\pi)d_{14}$ at 0° and 90°. In the parallel configuration the effective nonlinearity is maximized at 35.3° and is $d_{\mathrm{eff}}(\max) = (1/3)(2/\sqrt{3})(2/\pi)d_{14}$. In our experiment a wire grid polarizer was placed before the BaF_2 lens so that both input waves had the same polarization, at 45° from the [110] direction with $d_{\mathrm{eff}}(45°) = 0.97\, d_{\mathrm{eff}}(\max)$, which was 12% larger than for the orthogonal case.

In the orthogonal polarization configuration the incident pump-pulse energy was 5.9 μJ, or 65-kW peak power. A 5-cm focal-length BaF_2 lens focused the input beams to a 100-μm spot, giving a peak pump intensity of 410 MW/cm^2. The incident signal pulse energy was 1.76 μJ. The DBS GaAs was uncoated, resulting in 30% Fresnel reflection at each surface.

The homogeneity of the phase matching of the DBS GaAs crystal over its length was tested by

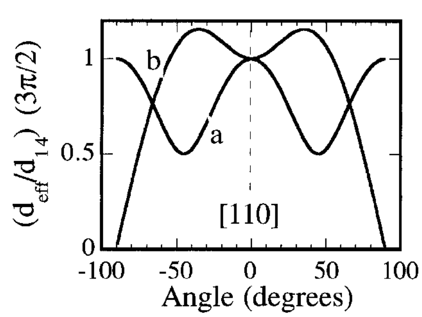

Fig. 3. Theoretical effective nonlinear coefficient d_{eff} as a function of the angle between the pump-beam polarization and the GaAs [110] direction (at normal incidence). For curve a, it is assumed that the input beams have the orthogonal polarizations; for curve b, that the polarizations are parallel. The normalization factor $(3\pi/2)$ is due to third-order quasi-phase matching.

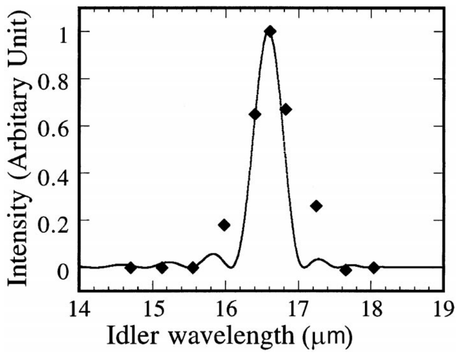

Fig. 4. Theoretical wavelength-tuning curve and measured data points. Optical loss was taken into account in the theoretical tuning curve. The inputs had linewidths of 10−15 cm^{-1}, comparable with the DBS GaAs acceptance linewidth of 16.4 cm^{-1}, so the measured tuning curve is broader than the theoretical prediction.

measurement of the wavelength-tuning curve. The input frequencies ω_p and ω_s were varied as $\omega_p + \omega_s = \omega_{laser}$, where ω_{laser} is the frequency of the 2.8-μm pump for the ZGP OPG. The wavelength-tuning curves for the two polarization states were similar. Figure 4 shows that the generated output was tunable from 15.6 to 17.6 μm. The FWHM of the measured wavelength-tuning curve was 29 cm^{-1}. With the DFG pump and signal linewidths of 10 cm^{-1} taken into account, the theoretical FWHM was 22 cm^{-1}, close to the observed value. The discrepancy may also be the result of sample period inhomogeneity.

The maximum idler conversion efficiency was 0.7% at 16.6 μm, close to the theoretical prediction obtained with a value of 83 pm/V for d_{14}.[6] If sample period inhomogeneity is taken into account, the theoretical prediction is still within the experimental error. Taking into account the Fresnel reflection, the measured external conversion efficiency corresponds to 2% internal conversion efficiency, or 5% internal quantum-conversion efficiency.

For the parallel configuration, the measured maximum conversion efficiency was a factor of 2 lower than that of the orthogonal configuration. This result is in accord with the theoretical prediction despite the fact that the effective nonlinearity is 1.12 times higher in the parallel case and the pump intensity is smaller by a factor of 2.3 owing to the 45° projection and 15% polarizer loss; thus the total predicted reduction in the DFG efficiency is approximately 2.1.

Higher conversion can be achieved with even shorter pulses at higher intensity. For a 1-ps pulse, the group-velocity walk-off distance for mixing 4.79- and 6.74-μm radiation is 11 mm, longer than our current crystal. In a separate test the surface-damage threshold of DBS GaAs was found to be 70 MW/cm^2 for 10-ns pulses. For 1-ps pulses, the surface-damage threshold should increase to more than 5 GW/cm^2, assuming scaling with the square root of pulse length, leading to a much higher conversion efficiency. With different input wavelengths and corresponding layer thickness, the interaction wavelengths can cover the entire GaAs transparency range of 2 to 17 μm.

In summary, we have fabricated 24-layer diffusion-bonded-stacked GaAs. The optical loss at long wavelengths is similar to that of single-crystal GaAs. The wavelength-tuning curve demonstrated that the bonding process maintained phase matching over the full crystal length of 6 mm. The peak external conversion was 0.7%, corresponding to 5% internal quantum-conversion efficiency at 16.6 μm. The generated mid-infrared radiation was tunable from 15.6 to 17.6 μm. With its large temperature, wavelength, and angular acceptance, this approach to engineered nonlinear optical materials appears promising for tunable infrared sources. We also expect to achieve much better results with first-order quasi-phase-matching GaAs. More details on DBS GaAs theory, fabrication, and optical experiments can be found in Ref. 7.

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